Electron attenuation lengths for free-electron-like metals

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Mean free paths for electrons in bulk jellium are calculated for hot electrons with energies from a few hundred to a few thousand electron volts and for values of r_s from 1.5 to 5. Exchange and correlation effects are included and the results are compared to relevant experiments.

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The experimental techniques of photoelectron spectroscopy and Auger electron spectroscopy are surface sensitive because of the small inelastic mean free path λ of electrons escaping from the solid. This quantity determines the average depth beneath the surface probed by these techniques and must be known accurately for a quantitative understanding of photoemission and Auger experiments.

We present a calculation of the mean free path in the energy range from a few hundred to a few thousand electron volts, and we also give an asymptotic formula for λ valid above a few thousand eV. A more detailed version of this work will be published elsewhere.[1] The calculation is valid for free-electron-like materials; we consider a material to be free-electron-like if its loss function $-\mathrm{Im}\, 1/\epsilon(q=0,\omega)$, as determined from characteristic-energy-loss experiments or from optical experiments, shows that the prominent loss mechanism is due to well defined plasmons which have an energy close to the free-electron value $\omega_p=(4\pi ne^2/m)^{\frac{1}{2}}$. Roughly speaking, materials which are not composed of transition or noble metal atoms (i.e., normal metals) are free-electron-like for our purposes. For such a material the inelastic mean free path due to valence-band excitations depends only on the energy of the hot electron and on the average electron density n of the material which is parameterized by $r_s=(3/4\pi n)^{\frac{1}{3}}(1/a_0)$ where a_0 is the Bohr radius.

Although the primary cause of the electron attenuation is due to the inelastic scattering of the hot electron by the valence electrons, the excitation of core electrons must also be considered as it typically reduces the mean free path by about 10% or so.

Previously, Quinn[2] and Kleinman[3] have calculated the mean free path for Al and Shelton[4] has carried out similar calculations of λ as a function of r_s up to hot-electron energy $\epsilon/\epsilon_f \leq 25$ where ϵ_f is the Fermi energy. We present here what we feel is a more accurate calculation than that of previous workers due to inclusion of exchange and correlation effects and we extend the calculations up to several thousand electron volts. Scattering due to surface plasmons is neglected; this is reasonable because of the rather large mean free paths at the higher energies of interest here and because at the surface there is some compensation between the decrease in bulk plasmon scattering and the increase in surface plasmon scattering.

The mean free path is given by[4,5]

$$\lambda(k)=\left(\frac{1}{\hbar}\frac{\partial E(k)}{\partial k}\right)\left(\frac{\hbar}{2|\Gamma(k)|}\right) \tag{1}$$

where $E(k)$ and $\Gamma(k)$ are the real and imaginary parts of the energy of an electron with momentum k and they are determined by

$$E(k)+i\Gamma(k)=\epsilon_k+M_k(E(k)+i\Gamma(k)) \tag{2a}$$

where the self-energy M_k is

$$M_k(\epsilon)=\frac{e^2}{2\pi^3}\int_0^{\epsilon-\epsilon_f} d(\hbar\omega) \times\int\frac{dq^3}{q^2}\frac{1}{\epsilon(q,\omega)}\frac{1}{\epsilon-\hbar\omega-\epsilon_{k-q}+i0^+} \tag{2b}$$

where $\epsilon_k=\hbar^2k^2/2m$, ϵ_f is the Fermi energy, and $\epsilon(q,\omega)$ is the dielectric function of the solid.

Equation (2a) can be solved approximately for $E(k)$ and $\Gamma(k)$ to obtain[5]

$$E(k)+i\Gamma(k)=\epsilon_0+\epsilon_k+Z(k)[M_k(\epsilon_k)-\epsilon_0] \tag{3a}$$

$$\epsilon_0=M_{k_f}(\epsilon_{k_f}) \tag{3b}$$

$$Z(k)=\left(1-\frac{\partial M_k(\epsilon)}{\partial\epsilon}\bigg|_{\epsilon=\epsilon_k}\right)^{-1}. \tag{3c}$$

Equation (2b) for the self-energy $M_k(\epsilon)$ is, of course, an approximation to the true self-energy $\mathfrak{M}$ which is determined from the system of equations

$$\mathcal{G}=g+g\mathfrak{M}\mathcal{G}, \tag{4a}$$

$$\mathfrak{M}=\mathcal{G}\Gamma V, \tag{4b}$$

$$V=v/\epsilon=v/(1-vP), \tag{4c}$$

$$P=\mathcal{G}\Gamma\mathcal{G}, \tag{4d}$$

where

$$\langle k|g|k\rangle=(\epsilon-\epsilon_k)^{-1}$$

and

$$\langle k|\mathcal{G}|k\rangle=(\epsilon-\epsilon_k-\langle k|\mathfrak{M}|k\rangle)^{-1}$$

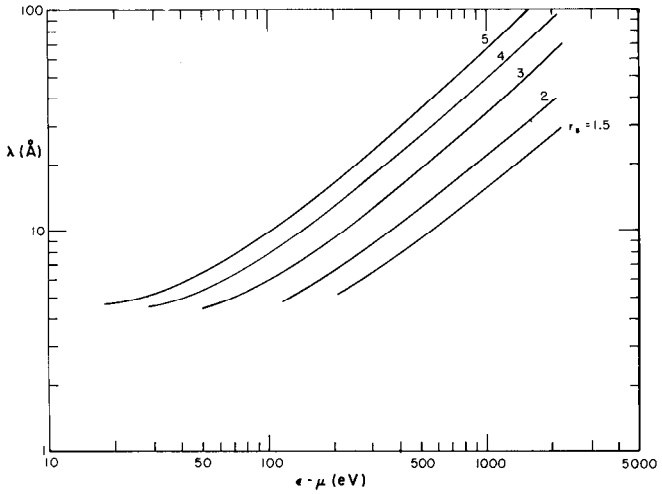

FIG. 1. The mean free path versus hot-electron energy measured with respect to the Fermi energy as determined by the "best" theory.

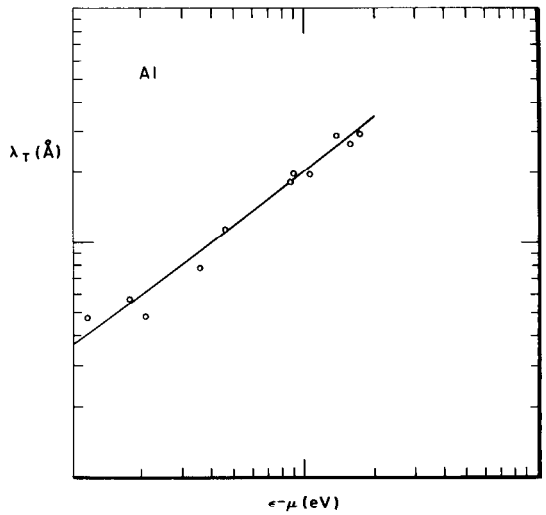

FIG. 2. The total mean free path for Al including inelastic scattering by both valence and core electrons is denoted by the solid line. The experimental points are due to Tracy.[9]

are the one-electron Green functions in the absence and presence of the Coulomb interaction v, $\mathfrak{M}$ is the self-energy, V is the effective electron–electron interaction, and Γ is the vertex correction. Equation (3) is obtained from Eq. (4) by setting $\Gamma=1$ and $\mathcal{G}=g$ in (4b).

Quinn[2] replaced Eq. (1) for $\lambda(k)$ by

$$\lambda_Q(k)=(\hbar k/m)(\hbar/|\mathrm{Im}M_k(\epsilon_k)|), \tag{5}$$

where the Lindhard dielectric function was used for ϵ in Eq. (2b). Shelton[4] retained Eq. (1) for $\lambda(k)$ and used the Lindhard dielectric function for ϵ in Eq. (2b) which corresponds to setting $\Gamma=1$ and $\mathcal{G}=g$ in Eq. (4d) as well as in Eq. (4b). We next obtain a more accurate result for $\mathfrak{M}$ by using a dielectric function in Eq. (2) which includes the effects of exchange and correlation. Singwi *et al.*[6] have developed a theory of the dielectric function which includes the effects of exchange and correlation but their theory does not yield a particularly tractable form for the dielectric function. We therefore turn to a simplified theory of electron correlation developed by Overhauser.[7] The theory employs a one-mode excitation spectrum; the excitations of the electron gas which consist of plasmons and electron–hole pairs are approximated by a single mode with a plasmon-like excitation spectrum[5] given by

$$\omega_q^2=\omega_p^2[1-G(q)]+\tfrac{1}{3}(\hbar k_f/m)^2q^2+(\hbar q^2/2m)^2, \tag{6a}$$

where

$$G(\bar{q})=0.275\bar{q}^2/(1+2.5\bar{q}^2+0.09375\bar{q}^4)^{\frac{1}{2}}, \tag{6b}$$

where $\bar{q}=q/k_f$ and the coupling constant for the electron–"plasmon" interaction is given by

$$g_q=[1-G(q)](e^2\hbar\omega_p^2/2q^2\omega_q)^{\frac{1}{2}}. \tag{7}$$

We use Eq. (7) in Eq. (2b) to obtain $M_k(\epsilon)$ and then the mean free path λ is found from Eqs. (3) and (1). In the case $G(q)=0$, determining the mean free path from Eqs. (7), (5), and (2) gives values for λ which agree with the "Quinn theory" to within 1% for $\epsilon_k/\epsilon_f\geq 9$ where ϵ_k is the energy of the hot electron. The calculation carried out by Shelton[4] is essentially equivalent to taking $G=0$ in Eq. (6a) and then determining λ from Eqs. (7), (2), (3), and (1).

We include correlation and exchange effects in the calculation of λ by using Eqs. (6) and (7) in determining M_K [as given by Eq. (2)]. The mean free path is then calculated from Eqs. (3) and (1). The results are shown in Fig. 1 where λ is plotted as a function of energy for various values of r_s.

The status of experimentally determined mean-free-path measurements has been reviewed by Powell.[8] As discussed by Powell, the experiments are difficult and tedious, and it is not easy to determine their accuracy. We will only refer to those experiments in which the mean free path has been measured by one worker for several values of hot-electron energy. Such experiments

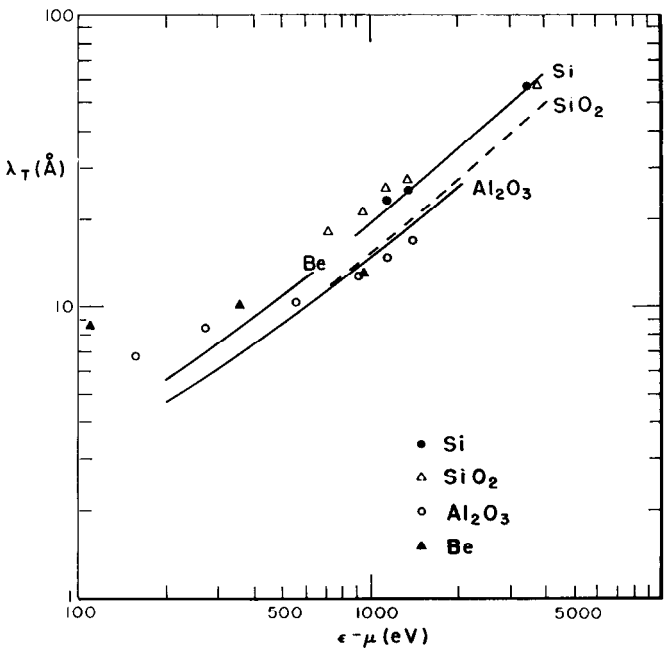

FIG. 3. The total mean free paths for Be,[10] Si,[11] SiO_2,[11] and Al_2O_3[12] are compared to experimentally determined points.

on free-electron-like materials have been carried out on Al,[9] Be,[10] Si,[11] SiO_2,[11] and Al_2O_3.[12]

The theory discussed above is incomplete in the sense that it neglects the inelastic scattering of hot electrons by the core electrons of the material under consideration. Powell[8] has worked out a theory for this effect and finds

$$\lambda_c(\epsilon_k) \simeq \frac{(2.55\times10^{-3})\epsilon_k A}{\rho \sum_i (N_i/\Delta E_i) \ln(4\epsilon_k/\Delta E_i)} \text{ (Å)}, \qquad (8)$$

where ϵ_k is the energy of the hot electron, A is the atomic (or molecular) weight of the component atoms or molecules of the material, ρ is the density, and ΔE_i is the average characteristic energy loss associated with the N_i core electrons. The effective mean free path λ_T is given by

$$\frac{1}{\lambda_T} = \left(\frac{1}{\lambda_c} + \frac{1}{\lambda}\right), \qquad (9)$$

where λ is the mean free path due to the valence electrons and given in Fig. 1. Typically λ_T is smaller than λ by roughly 10%. λ_T as determined from Fig. 1 and Eqs. (8) and (9) for Al is shown in Fig. 2 as are the experimentally determined points.[9] The agreement between theory and experiment is excellent; however, this must be in part fortuitous since the agreement is far better than could possibly be expected due to the uncertainties in both the theory and the experiment. In Fig. 3 we compare the results of the theory including core excitations with the experimental results for Be, Si, SiO_2, and Al_2O_3; the agreement is reasonable in all cases except SiO_2.

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